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May 12, 2006

PATENT APPLICATION
Attorney's Docket No.: 1021.2005-001

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Yi Hua Ma, Ivan P. Mardilovich and Erik E. Engwall

Application No.: 10/804,846 Group: 1724

Filed: March 19, 2004 Examiner: Frank M. Lawrence, Jr.

Confirmation No.: 7497

Title: Composite Gas Separation Modules Having Intermediate Porous Metal Layers

CERTIFICATE OF MAILING OR TRANSMISSION

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Ann M Gisso
Typed or printed name of person signing certificate

DECLARATION UNDER 37 C.F.R. § 1.132 OF YI HUA MA, Sc.D.

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Yi Hua Ma, Sc.D. of Worcester, MA, declare and state that:

1. I am a co-inventor of the subject matter described in U.S. Serial No. 10/804,846, claiming composite gas separation modules, methods for fabricating a composite gas separation module, methods for selectively separating hydrogen gas from a hydrogen gas-containing gaseous stream, hydrogen gas separators, methods of purifying hydrogen gas, and methods of manufacturing hydrogen gas separators. I also am a co-inventor of the subject matter described in U.S. Provisional Application No. 60/457,061, of which the subject application is claiming the benefit under 35 U.S.C. § 119. The subject patent application was filed on behalf of Worcester

Polytechnic Institute, 100 Institute Road, Worcester, Massachusetts 01609.

2. I have thoroughly studied the above-identified, subject patent application and its corresponding provisional application, and also studied the Office Action mailed from the U.S. Patent and Trademark Office on February 24, 2006 in the subject application.

3. I have over 30 years of experience in inorganic membrane and gas separation research. A summary of my professional biography appears as Attachment A.

4. A porous membrane is a thin layer containing pores of various shapes as shown in Fig. 1, where (A) shows cylindrical pores while (B) indicate conical pores. For effective separations, the pore size has to be small and is, in general, in the submicron range. Because of the small size of hydrogen molecules, porous membranes can not effectively separate hydrogen from other gas molecules [1,2].

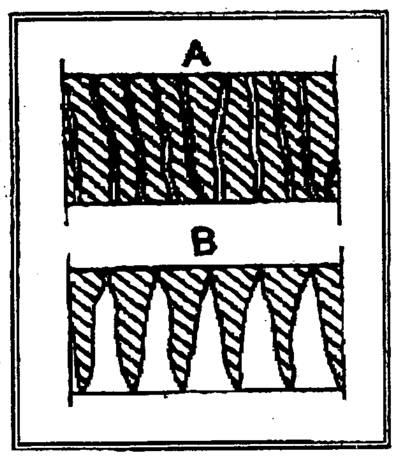

Fig. 1. Pores in a porous membrane (A) Straight cylindrical pores (B) Conical pores.

5. In the membrane research field the term "dense membrane" has become generally synonymous with a non-porous membrane. Inorganic membrane literature is replete with references which use the term "dense membrane" in this way. For example, as stated in "Fundamentals of Inorganic Membrane Science and Technology," Ed. Burggraff, A.J., et al.,

Elsevies (1996) in Chapter 11, written by Jose Sanchez, Ph.D. and Theodore P. Tsotsis, Ph.D. (who is the chair of the Chemical Engineering Department of the University of Southern California):

> 2. Dense Metal Membrane Reactor
>
> . . . .
>
> Pd together with a handful of other metals is permeable to hydrogen but virtually impermeable to other gases and, of course, liquids. The diffusion process through Pd, furthermore, is an activated process and at high temperatures such membrane show very reasonable permeance.

A copy of the relevant portion of this text appears as attachment B.

Another example of distinction between "dense metal membranes" and "porous metal membranes" can be found at page 126 of <u>Membrane Separation Systems, Recent Developments and Future Directions</u>, Baker, R.W., et al., Noyes Data Corporation (1991):

> 1.3 CERAMIC AND METAL MEMBRANES
>
> 1.3.1 Dense Metal Membranes
>
> Palladium and palladium alloy membrane can be used to separate hydrogen from other gases. Palladium membranes were extensively studied during the 1950's and 1960's and a commercial plant to separate hydrogen from refiner off-gas was installed by Union Carbide. [46,47] The plant used palladium/silver alloy membranes in the form of 25 μm-thick films.
>
> . . . .
>
> 1.3.2 Microporous Metal Membranes
>
> . . . The membranes have a tightly controlled pore size distribution and can be preduced with pore diameters ranging from 0.02-2.0 μm.

A copy of the relevant portion of this text appears as Attachment C.

Therefore, a dense membrane does not contain any pores and permeation is through the solution diffusion, whereby gas transport occurs when gas molecules first dissolve into a membrane and then diffuse across it. A dense palladium membrane is such a membrane, as are vanadium, tantalum and niobium.

6. One skilled in the relevant art would not consider porous intermediate layer (3) of 5,498,278 by Edlund (Edlund '278) to be a porous layer of hydrogen permeable material, nor would one skilled in the art consider porous intermediate layer (3) of Edlund '278 to be a dense hydrogen selective membrane.

7. One skilled in the relevant art would not consider the mesh layer of Bossard '542 to be porous layers of hydrogen permeable material, nor would one skilled in the art consider the mesh layers to be dense hydrogen-selective membranes.

8. One skilled in the relevant art would not consider the intermediate layer (9) of U.S. 2002/0020298 A1 by Drost et al. (Drost et al. '298) to be a porous layer of a hydrogen permeable material.

9. I hereby acknowledge that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Yi Hua Ma, Sc.D.

May 14, 2006

Date

BIOGRAPHICAL SKETCH

YI HUA MA

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Worcester Polytechnic Institute
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Professor Yi Hua Ma is a Professor of Chemical Engineering and Director, Center for Inorganic Membrane Studies at Worcester Polytechnic Institute. A chemical engineer by training, Professor Ma has been active in research involving inorganic membranes, inorganic materials, their adsorptive and transport properties in gases and liquids and industrial applications, including hydrogen production for fuel cell applications. He has published extensively with over 100 technical publications in the areas of inorganic membranes, adsorption and diffusion in porous inorganic adsorbents, mathematical modeling of transport processes and hydrogen separation and purification. Professor Ma has considerable administrative and project management experience. He was Head of the Chemical Engineering Department at WPI for 10 years between 1979 and 1989. He has been the Director of the Center for Inorganic Membrane Studies since 1987 and has managed the research collaboration between the Center staff and Center member companies including Alcoa, ExxonMobil Corporation, Texaco, United Technologies Corporation, and Amoco Chemical Company. Currently, he is collaborating with Shell E&P International, Inc. on a major project with funding over $2.5 million over a four-year period for hydrogen production using his patented palladium membrane technology. Professor Ma consults regularly for the US government and US and international corporations.

Education

Sc.D., 1967, Chemical Engineering, MIT, Cambridge, MA
M.S., 1963, Chemical Engineering, University of Notre Dame, Notre Dame, IN
B.S., 1959, Chemical Engineering, National Taiwan University, Taipei, Republic of China

Professional Experience

Worcester Polytechnic Institute, Department of Chemical Engineering
- Frances B. Manning Chaired Professor (2003-)
- Professor 1976-present; Department Head 1979-1989
- Director, Center for Inorganic Membrane Studies, 1988-present
- Associate Professor 1971-1976
- Assistant Professor 1967-1971

Honors and Awards

1994 WPI Board of Trustees' Award for Outstanding Research and Creative Scholarship

NASA Space Act Awards for the creative development of a technical innovation for reportable item MFS-26480 "Activation of Carbon Dioxide on Activated Carbon and

Attachment A"

Removal from Carbon through Direct Electrical Resistance Heating", September 3, 1997

AIChE Fellow

Consulting Experience

Procter & Gamble, 1994-1995
Foster-Miller, Inc., 1986-1996
Cabot Corporation, 1974-1986
US Army Natick Research and Development Laboratories, 1969-1980
NRG NuFnel Company, Newport Beach, CA 1974-1980

Professional Societies

President, Chinese-American Chemical Society, 2005-2006
Titular Member, IUPAC, 1999 - 2001
Editorial Board, Journal of Adsorption, 1993-
Editorial Board, Separation and Purification Technology, 1997-1999
International Advisory Board, Journal of Separations Technology 1990-1996
President, New England Association of Chinese Professionals, 1993-1995
Board of Directors, International Adsorption Society, 1992-1996
Vice President, International Zeolite Association, 1989-1992
Council Member, International Zeolite Association, 1986-1992
Member, American Institute of Chemical Engineers (AIChE)
Member, American Chemical Society
Member, Sigma Xi
Member, Adsorption and Ion Exchange Committee, AIChE

Field of Research

Adsorption and Diffusion in Zeolites and other Porous Materials, Nano Particle Synthesis, Separation Technologies, Interfacial Mass Transfer Inorganic Membrane Synthesis and Applications, Dense Membranes for Hydrogen and Oxygen separations, Membrane Reactors, Synthesis of Pillared Layered Clays,

Publications

Over 120 technical publications.

5867

MEMBRANE SEPARATION SYSTEMS

Recent Developments and Future Directions

by

R.W. Baker, E.L. Cussler, W. Eykamp,
W.J. Koros, R.L. Riley, H. Strathmann

NOYES DATA CORPORATION
Park Ridge, New Jersey, U.S.A.

Attachment "B"

Previous Page

1.3 CERAMIC AND METAL MEMBRANES

1.3.1 Dense Metal Membranes

Palladium and palladium alloy membranes can be used to separate hydrogen from other gases. Palladium membranes were extensively studied during the 1950s and 1960s and a commercial plant to separate hydrogen from refinery off-gas was installed by Union Carbide.[46,47] The plant used palladium/silver alloy membranes in the form of 25 μm-thick films. The technique used to make the membrane is not known, but films of this thickness could be made by standard metal casting and rolling technology. A number of problems, including long-term membrane stability under the high temperature operating conditions, were encountered and the plant was replaced by pressure-swing adsorption systems.

Small-scale systems, used to produce ultrapure hydrogen for specialized applications, are currently marketed by Johnson Matthey and Co. These systems also use palladium/silver alloy membranes, based on those developed by Hunter.[48,49] Membranes with much thinner effective palladium layers than were used in the Union Carbide installation can now be made. One technique that has been used is to form a composite membrane, with a metal substrate, onto which is coated a thin layer of palladium or palladium alloy.[50] The palladium layer can be applied by spraying, electrochemical deposition, or by vacuum methods, such as evaporation or sputtering. Coating thicknesses on the order of a few microns or less can be achieved. Johnson Matthey and Co. holds several patents covering this type of technology, including techniques in which composites are coated on both sides with palladium and then rolled down to form a very thin composite film.[51] The exact nature of the membranes used in their present systems is proprietary.

1.3.2 Microporous Metal Membranes

Recently, Alcan International[51,52] and Alusuisse[53,54] have begun to produce microporous aluminum membranes. The production methods used are proprietary, but both use an electrochemical technique. In the Alcan process aluminum metal substrate is subjected to electrolysis using an electrolyte such as oxalic or phosphoric acid. This forms a porous aluminum oxide film on the anode. The pore size and structure of the oxide film is controlled by varying the voltage. Once formed, the oxide film is removed from the anode by etching. The membranes have a tightly controlled pore size distribution and can be produced with pore diameters ranging from 0.02-2.0 μm.

1.3.3 Ceramic Membranes

The major advantage of ceramic membranes is that they are chemically extremely inert and can be used at high temperatures, where polymer films fail. Ceramic membranes can be made by three processes: sintering, leaching and sol-gel methods. Sintering involves taking a colloidal suspension of particles, forming a coagulated thin film, and then heat treating the film to form a continuous, porous structure. The pore sizes of sintered films are relatively large, of the order of 10-100 μm. In the leaching process, a glass sheet or capillary incorporating two intermixed phases is treated with an acid that will dissolve one of the phases. Smaller pores can be obtained by this method, but

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FUNDAMENTALS OF INORGANIC MEMBRANE SCIENCE AND TECHNOLOGY

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Laboratory des Materiaux et Procédes Membranes, (UMR 9987 CNRS-ENSCM-4411), Ecole Nationale Supérieure de Chimie 8 Rue de l'Ecole de Chimie, 34053 Montpellier, France

1996

ELSEVIER
Amsterdam — Lausanne — New York — Oxford — Shannon — Tokyo

"Attachment C"

TABLE 11.1
Types of membrane reactors

Acronym	Description
CMR	Catalytic membrane reactor
CNMR	Catalytic nonpermselective membrane reactor
PBMR	Packed-bed membrane reactor
PBCMR	Packed-bed catalytic membrane reactor
FBMR	Fluidized-bed membrane reactor
FBCMR	Fluidized-bed catalytic membrane reactor

is replaced by a fluidized bed the FBMR configuration results. The concept is really interesting but FBMRs have yet to gain widespread acceptance. In the CMR configuration the membrane provides both the separation and the reaction function. The concept, however, has found wider acceptance in the bioreactor area than with catalytic reactors. Finally, the PBCMR (and FBCMR) uses both a catalytic bed and a permselective membrane. This configuration would appear to be ideal for situations where a bifunctional catalytic function is desirable; we are not aware, however, of many examples of such PBCMR use.

Other reactor configurations and concepts have also been discussed in the technical literature. Most commonly cited are hybrid concepts, where the membrane reactor is used as an add-on stage to an already existing conventional reactor. This particular configuration has a number of attractive features, especially for applications involving conventional type porous membranes, which are characterized by moderate (Knudsen-type) permselective properties. Staged membrane reactors have received mention and so have reactors with multiple feed-ports and recycle. To facilitate the transport across the membrane in laboratory studies one often applies a sweep gas or a vacuum in the permeate side or a pressure gradient across the membrane. It is unlikely that the first two approaches, effective as they may be in laboratory applications, will find widespread commercial application.

11.2 DENSE METAL MEMBRANE REACTORS

The earlier membrane reactors involved the use of Pd and Pd alloy membranes. Pd together with a handful of other metals is permeable to hydrogen but virtually impermeable to other gases and, of course, liquids. The diffusion process through Pd, furthermore, is an activated process and at high temperatures such membranes show very reasonable permeances. The pioneering work on Pd membrane reactors was done by Gryaznov and coworkers in the former Soviet Union and some industrial groups in the U.S. and Europe. Gryaznov and

McGRAW-HILL DICTIONARY OF SCIENTIFIC AND TECHNICAL TERMS

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concentration of sensory and neural organs in the head. { ˌsef·ə·lə'zā·shən }

Cephalobaenida [INV ZOO] An order of the arthropod class Pentastomida composed of primitive forms with six-legged larvae. { ˌsef·ə·lō'bēn·ə·də }

Cephaloboidea [INV ZOO] A superfamily of free-living nematodes in the order Rhabditida distinguished by cephalic elaborations or ornamentations. { 'sef·ə·lə'bȯid·ē·ə }

Cephalocarida [INV ZOO] A subclass of Crustacea erected to include the primitive crustacean *Hutchinsoniella macracantha*. { ˌsef·ə·lō'kar·ə·də }

Cephalochordata [VERT ZOO] A subphylum of the Chordata comprising the lancelets, including *Branchiostoma*. { ¦sef·ə·lō,kȯr'dād·ə }

cephalodium [BOT] A small wart-like growth containing nitrogen-fixing cyanobacteria that is found on or in the thallus of some lichens with photobionts. { ˌsef·ə'lō·dē·əm }

Cephaloidae [INV ZOO] The false longhorn beetles, a small family of coleopteran insects in the superfamily Tenebrionoidea. { ˌsef·ə'lȯid·ē }

cephalomere [INV ZOO] One of the somites that make up the head of an arthropod. { sə'fal·ə,mir }

cephalometry [ANTHRO] The science of measuring the head, especially for determining the characteristics of a particular race, sex, or somatotype. { ˌsef·ə'läm·ə·trē }

cephalont [INV ZOO] A sporozoan just prior to spore formation. { 'sef·ə,länt }

cephalopelvic disproportion [MED] A condition in which the fetus is unable to pass safely through the pelvis during labor because of pelvic contraction, an unfavorable fetal position, or a large fetal head in relation to pelvic size. Abbreviated CPD. { ¦sef·ə·lō¦pel·vik ˌdis·prə'pȯr·shən }

Cephalopoda [INV ZOO] Exclusively marine animals constituting the most advanced class of the Mollusca, including squids, octopuses, and *Nautilus*. { ˌsef·ə'läp·ə·də }

cephalosporin [MICROBIO] Any of a group of antibiotics produced by strains of the imperfect fungus *Cephalosporium*. { ˌsef·ə·lə'spȯr·ən }

cephalothin [MICROBIO] An antibiotic derived from the fungus *Cephalosporium*, resembling penicillin units in structure and activity, and effective against many gram-positive cocci that are resistant to penicillin. { 'sef·ə·lə·thən }

cephalothorax [INV ZOO] The body division comprising the united head and thorax of arachnids and higher crustaceans. { ¦sef·ə·lə'thȯr,aks }

Cephalothrididae [INV ZOO] A family of ribbonlike worms in the order Palaeonemertini. { ˌsef·ə·lō'thrī·də,dē }

cephalotrichous flagellation [CYTOL] Insertion of flagella in polar tufts. { ˌsef·ə·lō'trī·kəs ˌflaj·ə'lā·shən }

Cepheid [ASTRON] One of a subgroup of periodic variable stars whose brightness does not remain constant with time and whose period of variation is a function of intrinsic mean brightness. { 'sē·fē·əd }

Cepheus [ASTRON] A constellation with right ascension 22 hours, declination 70°N. Abbreviated Cep. { 'sē·fē·əs }

Cephidae [INV ZOO] The stem sawflies, composing the single family of the hymenopteran superfamily Cephoidea. { 'sef·ə,dē }

Cephoidea [INV ZOO] A superfamily of hymenopteran insects in the suborder Symphyta. { sə'fȯid·ē·ə }

cepstrum [ACOUS] The Fourier transform of the logarithm of a speech power spectrum; used to separate vocal tract information from pitch excitation in voiced speech. { 'sep·trəm }

cepstrum vocoder [ENG ACOUS] A digital device for reproducing speech in which samples of the cepstrum of speech, together with pitch information, are transmitted to the receiver, and are then converted into an impulse response that is convolved with an impulse train generated from the pitch information. { 'sep·trəm 'vō¦kōd·ər }

Ceractinomorpha [INV ZOO] A subclass of sponges belonging to the class Demospongiae. { sə¦rak·tə·nə'mȯr·fə }

ceramagnet [ELECTROMAG] A ferrimagnet composed of the hard magnetic material $BaO{\cdot}6Fe_2O_3$. { 'se·rə,mag·nət }

ceramal *See* cermet. { sə'ram·əl }

Cerambycidae [INV ZOO] The longhorn beetles, a family of coleopteran insects in the superfamily Chrysomeloidea. { se·rəm'bī·sə,dē }

cerambycoid larva [INV ZOO] A beetle larva that is morphologically similar to a caraboid larva except for the former's absence of legs. { sə'ram·bə,kȯid 'lar·və }

ceramet *See* cermet. { sə'ram·ət }

ceramic [MATER] **1.** Inorganic, nonmetallic materials processed or used at high temperature, generally including oxides, nitrides, borides, carbides, silicides, and sulfides. Intermetallic compounds such as aluminides and beryllides are also considered ceramics, as are phosphides, antimonides, and arsenides. **2.** Consisting of such a product. { sə'ram·ik }

ceramic aggregate [MATER] **1.** Portland cement concrete containing lumps of ceramic material. **2.** Concrete made with porous clay to reduce its weight. { sə'ram·ik 'ag·rə·gət }

ceramic amplifier [ELECTR] An amplifier that utilizes the piezoelectric properties of semiconductors such as silicon. { sə'ram·ik 'am·plə,fī·ər }

ceramic-based microcircuit [ELECTR] A microminiature circuit printed on a ceramic substrate. { sə'ram·ik,bāst 'mī·krō,sər·kət }

ceramic capacitor [ELEC] A capacitor whose dielectric is a ceramic material such as steatite or barium titanate, the composition of which can be varied to give a wide range of temperature coefficients. { sə'ram·ik kə'pas·əd·ər }

ceramic cartridge [ENG ACOUS] A device containing a piezoelectric ceramic element, used in phonograph pickups and microphones. { sə'ram·ik 'kär·trij }

ceramic coating [MET] A nonmetallic, inorganic coating made of sprayed aluminum oxide or of zirconium oxide, or a cemented coating of an intermetallic compound, such as aluminum disilicide, of essentially crystalline nature, applied as a protective film on metal to protect against temperatures above 1100°C. { sə'ram·ik 'kōd·iŋ }

ceramic earphones *See* crystal headphones. { sə'ram·ik 'ir,fōnz }

ceramic fiber [MATER] A small-dimension filament or thread composed of a ceramic material, usually alumina and silica, used in lightweight units for electrical, thermal, and sound insulation, filtration at high temperatures, packing, and reinforcing other ceramic materials. { sə'ram·ik 'fī·bər }

ceramic filter [ELECTR] A type of mechanical filter that uses a series of resonant ceramic disks to obtain a band-pass response. { sə'ram·ik 'fil·tər }

ceramic glaze [ENG] A glossy finish on a clay body obtained by spraying with metallic oxides, chemicals, and clays and firing at high temperature. { sə'ram·ik 'glāz }

ceramicite [PETR] A porcelained pyrometamorphic rock composed of basic plagioclase and cordierite with a small amount of hypersthene and a groundmass of glass. { sə'ram·ə,sīt }

ceramic magnet [ELECTROMAG] A permanent magnet made from pressed and sintered mixtures of ceramic and magnetic powders. Also known as ferromagnetic ceramic. { sə'ram·ik 'mag·nət }

ceramic microphone [ENG ACOUS] A microphone using a ceramic cartridge. { sə'ram·ik 'mī·krə,fōn }

ceramic mold casting [MET] A precision casting process using a ceramic body fired to high temperature as the mold, and carbon, low-alloy, or stainless steel as the casting. { sə'ram·ik 'mōld ˌkast·iŋ }

ceramic pickup [ENG ACOUS] A phonograph pickup using a ceramic cartridge. { sə'ram·ik 'pik·əp }

ceramic radiant [ENG] A baked-clay component of a gas heating unit which radiates heat when incandescent from the gas flame. { sə'ram·ik 'rād·ē·ənt }

ceramic reactor [NUCLEO] A nuclear reactor in which the fuel and moderator assemblies are made from high-temperature-resistant ceramic materials such as metal oxides, carbides, or nitrides. { sə'ram·ik rē'ak·tər }

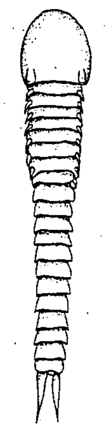
ceramic rod flame spraying [MET] A method of flame spraying in which the ceramic rod is fed into a gun that utilizes an oxyfuel gas flame to atomize and airblast the rod material to the substrate. { sə'ram·ik 'räd ˌflām ˌsprā·iŋ }

ceramics [ENG] The art and science of making ceramic products. { sə'ram·iks }

ceramic tile [MATER] A burned-clay product composed of a clay body with a decorative surface glaze; used principally for decorative and sanitary effects. { sə'ram·ik 'tīl }

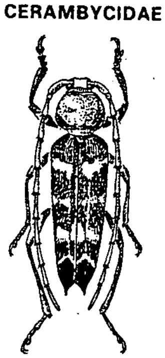
ceramic tool [DES ENG] A cutting tool made from metallic oxides. { sə'ram·ik ˌtül }

CEPHALOCARIDA



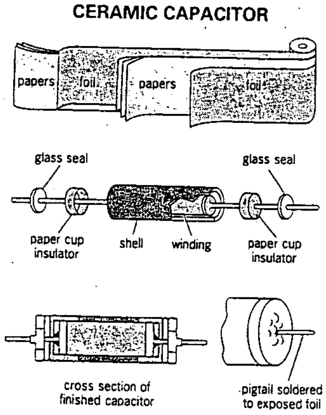
Hutchinsoniella macracantha.

CERAMBYCIDAE



A longhorn beetle. *(From T. I. Storer and R. L. Usinger, General Zoology, 3d ed., McGraw-Hill, 1957)*

CERAMIC CAPACITOR



Ceramic capacitor constructed in chip form, showing cutaway of finished chip.